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Letter to the Editors

Hardening of Fe–Cu alloys at elevated temperatures by electron and neutron irradiations

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Abstract

Comparative experiments using 2.5 MeV electron and neutron irradiations to Fe–Cu model alloys have been undertaken to study the gamma-ray induced hardening of RPV steels. Hardness changes induced by Cu precipitates for Fe–0.6%CU model alloy was confirmed. The difference between electron and neutron irradiation hardening was very small on a dpa basis except for very low dpa. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

Fast neutron fluence ($E > 1$ MeV) has been mainly used as a scaling parameter for irradiation embrittlement of reactor pressure vessel steels (RPVs). RPVs are exposed to not only fast neutrons but also thermal neutrons and gamma-rays. Although gamma-rays can produce radiation damage through Compton scattering and pair production, dpa cross-sections due to their processes are very small as compared with that by fast neutrons. The contribution of gamma-rays to dpa in RPVs is several % in commercial reactors (PWR: ~1%, BWR: ~3%) [1]. Therefore, it has been considered that only fast neutron damage dominates the irradiation embrittlement.

However, there is a special instance that shows a gamma-ray effect. A series of reevaluation on the high flux isotope reactor (HFIR) surveillance tests has revealed that the unexpected embrittlement observed in surveillance test materials is due to the high-level gamma-ray irradiation field [2]. Furthermore, there is an estimation for an advanced BWR that over 30% of the total dpa experienced by the vessel is caused by gamma-rays because the contribution to dpa from gamma-rays increases substantially with increasing water gap [3]. In such a case, gamma-ray effects cannot be ignored, obviously.

Another important point that is open to argument is whether or not gamma-rays can be much more effective for embrittlement on dpa basis than fast neutrons. In ion-irradiated NiSi and CuAu alloys, the amount of irradiation-induced segregation is larger for lighter ions even at the same calculated dpa. This experimental result has been explained as due to the difference in freely migrating defect (FMD) production rate [4,5]. Gamma-rays are expected to be much more effective than fast neutrons for producing FMD. If FMD plays an important role also in irradiation-induced embrittlement in RPVs, even a small dose of gamma-rays cannot be ignored.

Therefore, the following point has to be discussed: whether or not gamma-rays cause embrittlement more effectively than fast neutrons on a per dpa basis. Considerations of gamma-ray effects are essential for a more complete understanding and more accurate prediction of irradiation embrittlement of RPV steels.

The purpose of the present study is to evaluate the efficiency of gamma-ray-induced hardening experimentally by comparing the irradiation hardening caused by high-energy electrons with that by neutrons. Gamma-rays are known to generate atomic displacements in medium-Z metals indirectly through the production of energetic electrons and positrons primarily via Compton scattering and/or pair production interaction. Hence, we can simulate the effect of Gamma-rays on the hardening by using electron irradiation. It is generally recognized

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that two types of radiation-induced microstructural features contribute to hardening in RPV steels containing Cu as an impurity element; matrix damage and Cu-rich precipitates [6]. There have already been reported the comparisons of irradiation hardening by electron and neutron irradiations for iron-based alloys [7,8]. However, the difference in effects between electron and neutron irradiations, and hence between gamma-ray and neutron irradiations on RPV steels has not been fully clarified. Main difficulty of the previous comparisons lies in the fact that total dpa and dpa rates between the electron and neutron irradiations do not coincide in the previous experiments. In this paper, we report the result that shows the clear comparison through the series of systematic irradiation experiments.

2. Experimental procedure

Materials used in the present experiment were two kinds of Fe–Cu model alloys with high Cu (0.6 wt% Cu) and low Cu (0.02 wt% Cu) contents. We use Fe–0.02%Cu alloy as a reference to investigate the Cu effect. The concentration of impurities other than Cu was very low; (C: 0.002, Si: 0.002, O: 0.015, N: 0.0006 (wt%)). Before irradiation, both materials were heat-treated at 850 °C for 2 h and then were quenched to water. Specimens with 10 mm × 10 mm × 1 mm in dimension were irradiated with 2.5 MeV electrons using a Dynamitron accelerator at Takasaki Establishment of Japan Atomic Energy Research Institute (JAERI-Takasaki). The dpa rate of the irradiation was 6.5×10^{-9} dpa/s, and the specimen temperatures were controlled at 250 °C during irradiation. To study the effect of irradiation temperature, some specimen was irradiated at 275, 290 or 320 °C. Other specimens were irradiated at 250 °C at the Japan Materials Testing Reactor (JMTR) and the Kyoto University Reactor (KUR). The dpa rate for both irradiations was 9×10^{-9} dpa/s. For low dose neutron irradiation, the Yayoi Reactor of the University of Tokyo was also used. In this case, however, the irradiation temperature was 240 °C and the dpa rate was 8×10^{-11} dpa/s.

We measured Vickers hardness (Hv) before and after irradiation as an index of irradiation embrittlement. The applied load was 500 gf and the measuring temperature was room temperature (about 25 °C). Small angle neutron scattering (SANS) measurements were also performed for some of the specimens using the SANS-J facility at the Japan Research Reactor (JRR-3M). The details of the result on SANS measurements will be reported elsewhere [9].

The dpa distribution for the electron irradiation as a function of depth was calculated using the Monte Carlo simulation code, modified EGS4 (Electron–Gamma Shower 4) [10], where the cross-section of Mckinley and Feshback was used as the scattering cross-section, and the displacement energy of Fe atom was assumed to be 40 eV [11]. The indentation depth and accompanying plastic deformation area are considered to be less than 75 μm from the surface. Within this area, the cross-section for the defect (Frenkel pair) production does not depend on the depth and is about 3.4×10^{-27} m^2. The dpa for neutron irradiation was calculated using the dpa cross-section in accordance with ASTM recommendation [11]. When we calculated them, the actual neutron energy spectrum of each reactor was carefully taken into account. It should be noted here that the irradiation fields of these reactors have already been well characterized.

3. Results and discussion

Fig. 1 shows the electron fluence dependence of irradiation hardening for Fe–0.6%Cu and Fe–0.02%Cu alloys. Each data point represents the mean of 10 measurements. As shown in the figure, the change in hardness by the irradiation in the Fe–0.6%Cu alloy tends to be saturated at the high fluence. By contrast, irradiation hardening in the Fe–0.02%Cu alloy is much smaller than that in Fe–0.6%Cu alloy, and increases linearly as a function of electron fluence. SANS measurements have revealed that Cu precipitates are formed in Fe–0.6%Cu alloy after irradiation. The radius and the number density of Cu precipitates in Fe–0.6%Cu alloy irradiated to the fluence of 2.2×10^{23} e/m^2 are estimated to be 3.8 nm and 5×10^{22} m^{-3}, respectively. In Fe–0.02%Cu alloy, such precipitates could not be observed. Thus, the results clearly shows that the hardening in Fe–0.6%Cu alloy is caused mainly by Cu precipitates, while in Fe–0.02%Cu

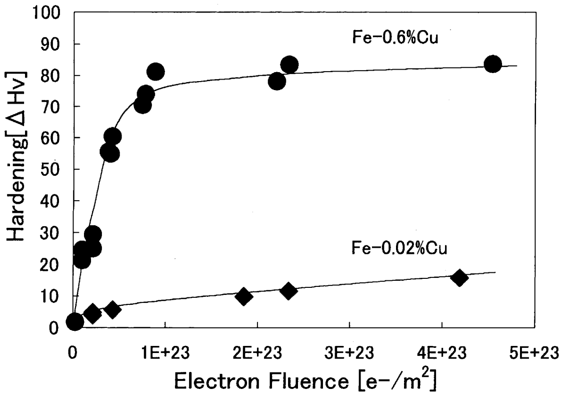

Fig. 1. Change in hardness as a function of electron fluence for Fe–0.6%Cu and Fe–0.02%Cu alloy. Irradiation temperature is 250 °C. Each hardening data represent the mean of 10 measurements.

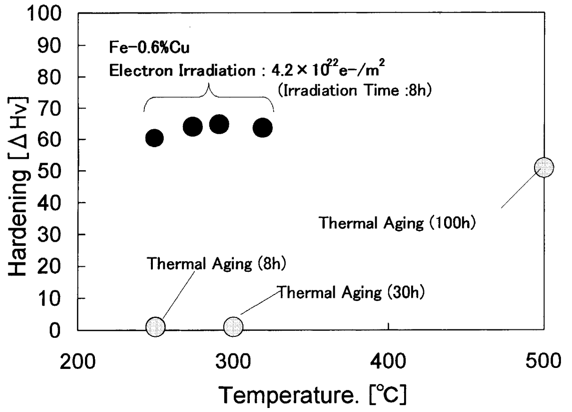

Fig. 2. Irradiation temperature dependence of hardening for Fe–0.6%Cu alloy. Thermal aging data are also plotted.

alloy, the hardening is caused only by matrix damage. Fig. 2 shows the irradiation temperature dependence of hardening for the Fe–0.6%Cu alloy. The hardening is almost the same in the temperature range from 250 to 315 °C. The results of thermal aging are also shown in the figure. The figure clearly shows that the hardening is not caused only by the thermal aging. The change in hardness in the Fe–Cu alloys is, therefore, definitely caused by the electron irradiation.

In Fig. 3(a), we compare the results for electron and neutron irradiation-induced hardening. The dpa rates for JMTR and KUR irradiations are almost the same within a factor of two to that for electron irradiation. However, the dpa rate for the Yayoi reactor was 100 times lower than that for the electron irradiation. For Fe–Cu model alloys, Hasegawa et al. [12] have verified the −1/4 power dependence on dpa rate with regard to irradiation hardening from the ion-irradiation experiments; i.e., $\Delta Hv \propto (\text{dpa rate})^{-1/4}$. To correct the effect of dpa rate, we have used this dependence. Fig. 3(b) shows the irradiation hardening versus dpa after the dpa rate correction. All the irradiation hardening data are corrected to the values for the dpa rate of 6.5×10^{-9} dpa/s, which is the average value for electron irradiation. Even after the rate effect correction, the dpa dependence of the hardening is almost the same for electron and neutron irradiations, except for very low dpa. Namely, the irradiation-induced hardening in Fe–Cu alloys can be scaled with the calculated dpa for electron and neutron irradiations. This result apparently implies that energy transferred elastically from the irradiating particles to the target Fe–0.6%Cu alloy dominates the Cu precipitation process and that we do not need to consider the defect annihilation during the cascade quench or the temperature dependence of surviving defect fraction (FMD efficiency) [4,5,13] as important factors for the description of the phenomenon. This may be due to the special situation of the present alloy system; a large number of Cu atoms are solved supersaturatedly in the specimen, and the irradiation forces the system easily to the thermally equilibrium state. For the Fe–0.02%Cu alloy, the hardness change due to the irradiation was very small, and the comparison for scaling data is not feasible.

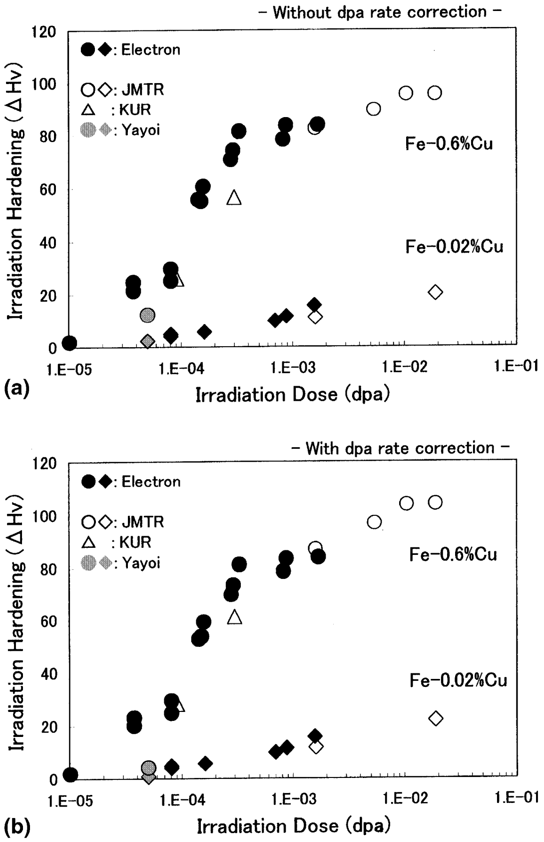

Fig. 3. (a) Comparison of electron and neutron irradiation hardening. (b) Same as Fig. 3(a) except that all data are corrected to values corresponding to the dpa rate of 6.5×10^{-9}.

4. Summary

Comparative experiments using electron and neutron irradiations to Fe–Cu model alloys have been undertaken to study the gamma-ray-induced hardening of RPV steels. Hardness changes induced by Cu precipitates for Fe–0.6%Cu model alloy was confirmed. The difference between electron and neutron irradiation hardening was very small on a dpa basis except for very low dpa. Thus, both gamma-ray and neutron-induced hardening can be well scaled with calculated dpa.

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